FULL-SIZED PLATES IRRADIATION WITH HIGH UMo FUEL LOADING - FINAL RESULTS OF IRIS 1 EXPERIMENT -

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ABSTRACT

As a part of the French UMo Group qualification program, IRIS 1 experiment contained full-sized plates with high uranium loading in the meat of 8 g.cm^{-3}. The fuel particles consisted of 7 and 9 wt% Mo-uranium alloys ground powders. The plate were irradiated at OSIRIS reactor in IRIS device up to 67.5% peak burnup within the range of 136 W.cm^{-2} for the heat flux and 72°C for the cladding temperature.
After each reactor cycle the plates thickness were measured. The results show no swelling behaviour differences versus burnup between UMo7 and UMo9 plates. The maximum plate swelling for peak burnup location remains lower than 6%.
The wide set of PIE has shown that, within the studied irradiation conditions, the interaction product have a global formulation of "(U-Mo)Al_{-7}" and that there is no aluminium dissolution in UMo particles.
IRIS1 experiment, as the first step of the UMo fuel qualification for research reactor, has established the good behaviour of UMo7 and UMo9 high uranium loading full-sized plate within the tested conditions.

1 Introduction

The French Program for UMo fuel qualification has been launched in 1999 in close collaboration with five partners : CERCA, CEA, FRAMATOME-ANP, TECHNICATOME and COGEMA. The aim of this program is to contribute to the development of a high performance and reprocessable UMo fuel. To reach this goal, the French group choose to irradiate full sized experimental plates with low enriched uranium (^{235}U <20%) and high uranium loading up to 8 g.cm^{-3}[1].
The first step of this program was to irradiate three fuel full-sized plates in OSIRIS reactor up to 241 Full Power Days (FPD) from September 1999 to January 2001.

2 Full-sized plates fabrication and characterization

The three low enrichment uranium plates irradiated in IRIS device were supplied by CERCA [2]. Two molybdenum contents in UMo alloy were tested : 7.6 wt% for one plate and 8.7wt% for the two others.
As for UMUS experiment[3], UMo powders were produced by grinding alloys ingots. As previously seen, this process leads to a powder of irregular morphology with high oxygen content and a rather high porosity level in the plate meat, up to 13%. The volume fraction of fuel particles was greater than 50%, so that the uranium densities were up to 8.3 g.cm^{-3}.

The results of the fuel plate inspections, based on metallographic examinations, X-Ray diffraction, blister test, … indicated that the plates conformed the specifications [2].
The main characteristics of the 3 plates are summarized in Table 1 and Fig. 1 shows the microstructure of a an as-fabricated plate.

Plate	**Ref.**	**Mo/UMo (wt%)**	**^{235}U/U (%)**	**U_{total} (g)**	**$^{235}U_{total}$ (g)**	**U density ($gU.cm^{-3}$)**	**Mean plate thickness (mm)**	**Porosity (%)**
#1	U7MQ2003	7.6	19.65	150.4	29.6	8.3	1.30	12.4
#2	U9MQ2051	8.7	19.67	145.6	28.6	7.9	1.29	13.0
#3	U9MQ2053	8.7	19.67	145.5	28.6	8.1	1.29	10.9

Table 1 : Characteristics of IRIS 1 experiment UMo plates

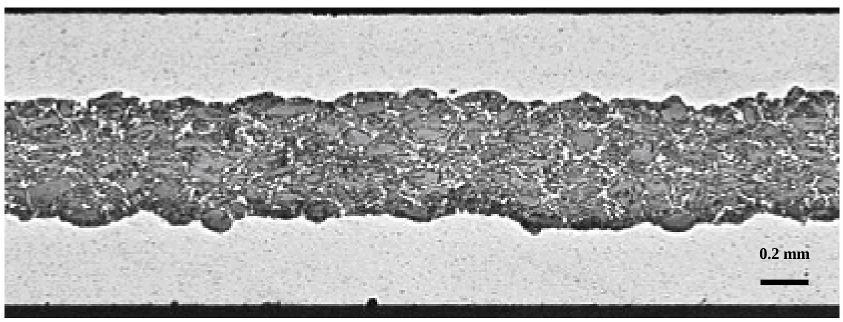


Fig. 1 : Metallographic section of as fabricated UMo fuel plate

3 Irradiation test conditions

The irradiation of the three plates was carried out in IRIS[4] device placed in a OSIRIS reactor in-core peripheral position.
IRIS device is associated to an in-pool plate thickness measurements device. The measurements are performed after each reactor cycle. Four plates can be placed in the device, but only two places were used for UMo experiment IRIS1.
The irradiation started with plates P#1 and P#3. Therefore, after, the second cycle, the plate P#3 was slightly warping probably during a handling. So it was decided to replace it by new one with similar characteristics (P#2).
IRIS1 experiment irradiation conditions and burnup are summarized in Table 2.

	U7MQ2003 P#1	**U9MQ2051 P#2**	**U9MQ2053 P#3**
Irradiation :			
Full power Day	241	155	46
Burnup (calculated)			
Plate average	47	35	13
Peak	67.5	55	22
Calculated values			
Peak clad. Temp. (°C)	69	72	74
Peak heat flux ($W.cm^{-2}$)	124	136	145

Table 2 : Burnup, peak cladding temperature and heat flux of plates from IRIS1 experiment

4 Post-irradiation examinations

4.1 Non destructive examinations

4.1.1 Visual inspection

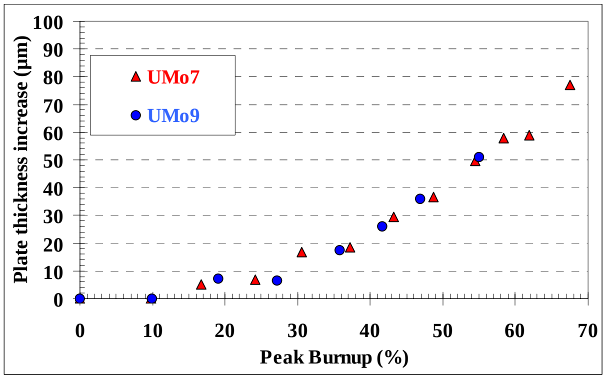
A complete visual inspection of the most irradiated fuels plates (P#1 and P#2) was done. Both plates presented no evidence of any kind of deformation, blister or oxide spallation.
The light grey oxide layers were founded to be uniform and to be localised in the meat zone.

4.1.2 Gamma scanning

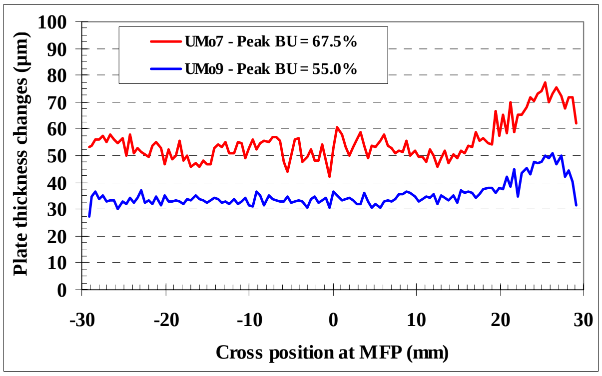
Gross and quantitative gamma scanning of plates P#1 and P#2 were done. For both kind of analyse, axial and transverse profiles were measured.
Gross gamma scanning measurements are representative of the whole irradiation average plate power . So they were used to access to the burnup relative profiles.
The location of the samples for destructive examinations and the peak burnup was determined from ^{137}Cs quantitative analysis.

4.1.3 Thickness measurements

The plate thickness changes were checked after each reactor cycle by the measurement of five axial and one transversal profiles. These latest were done at the maximum flux plan (MFP).
Fig. 2-a shows the evolution of the maximum plate thickness increase during irradiation.
The plates thickness increase profiles at MFP are given Fig. 2-b for both plates. The profiles shapes are consistent with the burnup profiles deduced from gamma scanning measurements.
At the end of irradiation, peak plate thickness increases are 77 µm (5.9 %) and 51 µm (4,4 %) respectively for UMo7 and UMo9 plates.



a/



b/

Fig. 2 : Maximum plate thickness increases versus burnup (a) End irradiation plate swelling profiles at MFP (b)

4.2 Destructive examinations

Destructive post-irradiation examinations have so far concentrated on detailed analysis of the most irradiated plate (UMo7 P#1). On UMo9 plate P#2, only metallographic examinations at peak burnup location were done for comparison.

4.2.1 Sampling procedure

Three 15.4 mm ϕ circular samples were drilled in the UMo7 plate. Their positions are drawn Fig. 3.

Two of them were taken at MFP (n° 1 & 2) and the third one was taken in a coldest part corresponding to the coolant entrance side. For comparison, only sample n°1 was drilled in UMo9 plate.

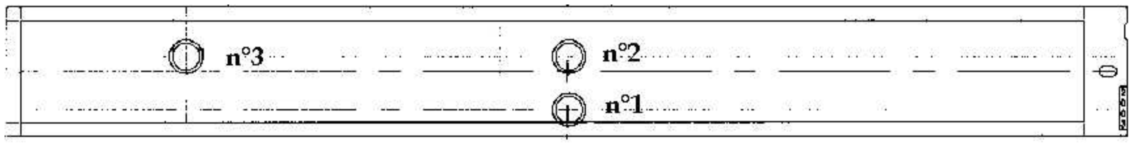
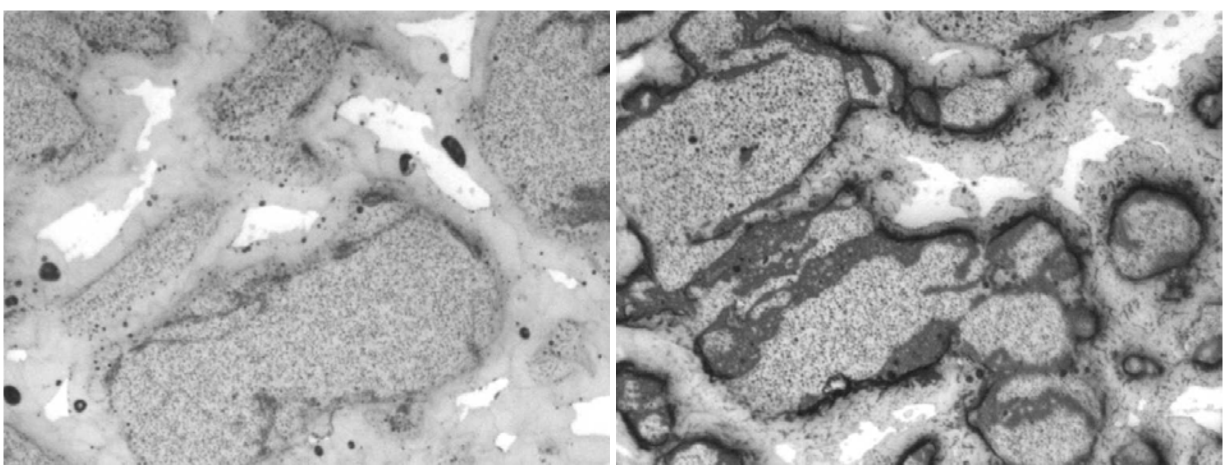


Fig. 3 : Samples location on UMo7 plate

Sample n° 1 corresponds to the maximum burnup location. Two areas of this sample have been examined : one at the edge plate side (area n° 1-A) and the other at the centre plate side (n° 1-B).

4.2.2 Fuel Microstructure

All the three samples exhibit almost the same microstructure. A typical view of it is given Fig. 4-a, with a little fraction of residual aluminium, some large pores remaining from initial porosity, high density of fine bubbles inside UMo fuel particles, and large areas of an interaction product layer without bubbles.
There is only one singular area (see Fig. 4-b), less than 1 mm width, located in area n°1-A which corresponds to the peak burnup location, but limited temperature due to the proximity of the plate side. In this area, bubbles are also detected in the UMo-Al interaction layer, and the dark areas, corresponding to the UMo powder oxidation induced by grinding process, are thicker.
No significant differences were observed for UMo9 plate microstructure.



a/ 20 µm b/ 20 µm

Fig. 4 : UMo7 microstructure at MFP : a/ middle of the plate (BU=61%) - b/ edge of the plate(BU=67.5)

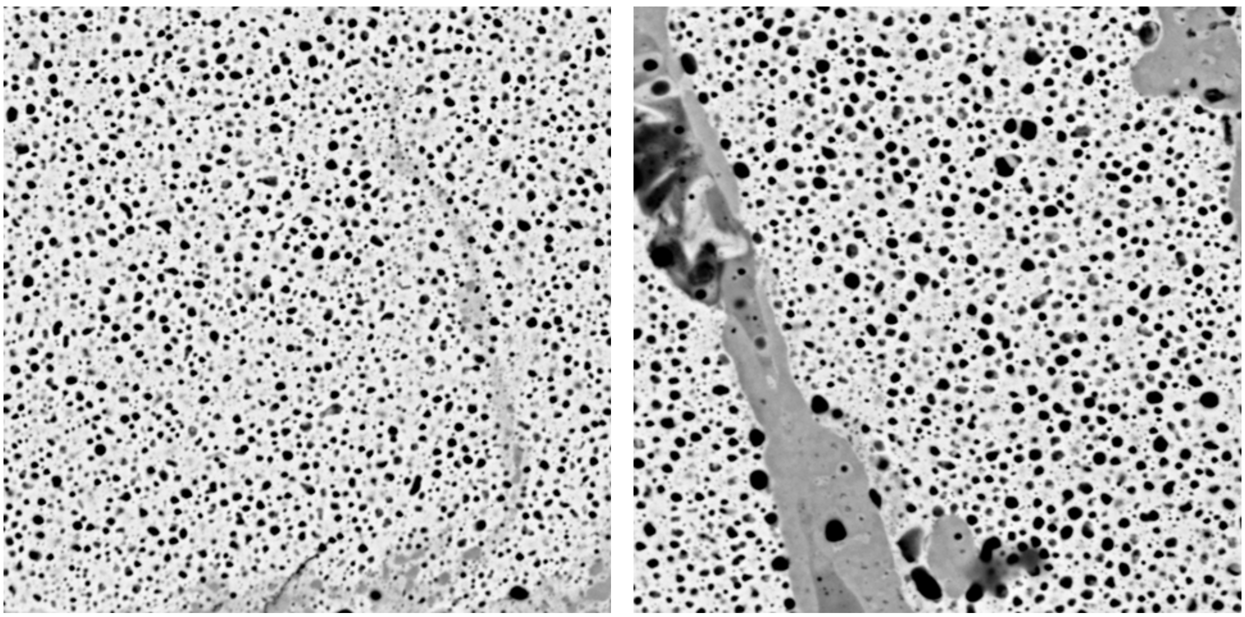
Quantitative image analyses were done in order to evaluate the different phase proportions. The results are given Table 3. They indicate that :

- As-fabricated porosity ("Large pores") decreases from l2.3% to less than 2.6%
- A large amount of aluminium was consumed by reaction with the UMo fuel. Indeed, aluminium content decreases from 35.5% to about 5% while the interaction compound volume rises up to more than 35%.
- The UMo fuel fraction has increased.
- Interaction layer thickness ranges from 4 to 6 µm
- The average oxide thickness on the plate surface is almost the same for all the samples. It remains below 16 µm

Sample	BU (%)	UMo (%)	interaction layer (%)	Al (%)	large pores (%)	Int. layer thickness (µm)	Plate oxide thickness (µm)
as fabricated✍	0	52.2	0	35.5	12.3	0	0
N° 1-a	67.5	56 ± 1	36 ± 1	5.5 ± 0.5	2.6 ± 0.5	~5	15
N°1-b	61.0	52± 1	42 ± 1	4.5 ± 0.5	1.1 ± 0.5	~6	15
N°2	58.0	58± 1	37 ± 1	3.9 ± 0.5	1.1 ± 0.5	~5.5	14
N° 3	41.7	57 ± 1	37 ± 1	5.7 ± 0.5	1 ± 0.5	~4	16

Table 3 : Phase volume proportions in UMo7 plate : as-fabricated value and quantitative image analyses.

A major gaseous fission product precipitation is observed in the UMo fuel particles. Metallographic examinations clearly point out the fact that the bubbles density evolves with the local burnup.



a/ UMo7 –BU=61% (n°1-b) 5 µm b/ UMo7- BU=67.5% (n°1-a) 5 µm

Fig. 5 : BSE images of UMo7 particles at MFP (sample n°1) a/ Plate center side (61% BU) – b/ Plate edge side (67.5% BU)

SEM examinations were done in order to obtain a quantitative evaluation of the bubble sizes and their repartition. Back scattered electrons images of UMo7 particles at MFP are given **Fig. 5** for two burnup.

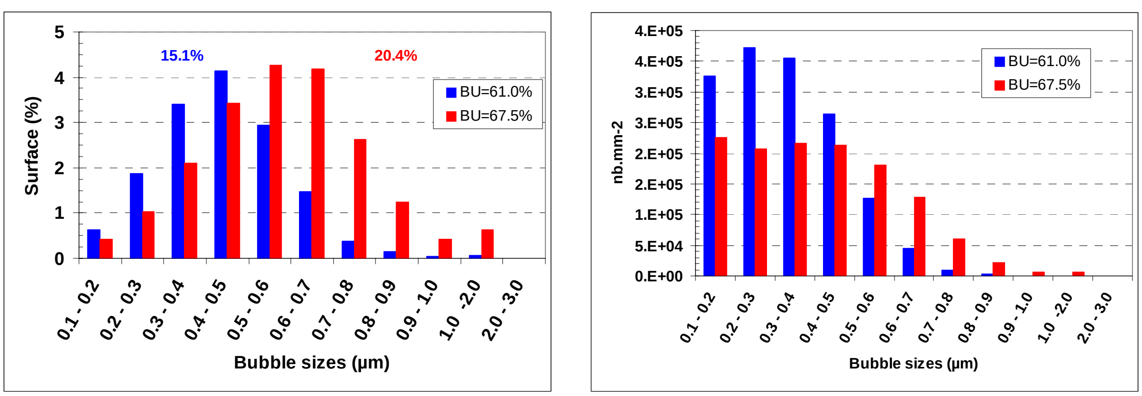


Fig. 6 : Fission gas bubble sizes repartition for BU = 61 and 67.5%

✍ calculated from the immersion volume of the plate and material theoretical densities

It can be seen that for BU from 61 to 67.5%, fission gas bubbles morphology doesn't change. It remains almost spherical and no indication of abnormal interlink can be seen. From 61 to 67.5% burnup, bubble's volume in the UMo7 particles rises up from 15.1 to 20.4% (see **Fig. 6**) mainly due to a slight increase of the diameter of the bubbles. However the very uniform distribution, with bubble sizes of less than 1 µm, that show no tendency to interlink, confirms the stable swelling behaviour of UMo particles for these samples.

4.2.3 Electron probe microanalysis (EPMA)

EPMA X-ray mappings were done in order to know the major components repartitions (see Fig. 7). Fission products accumulation is observed at the interface between the interdiffusion phase and the aluminium.

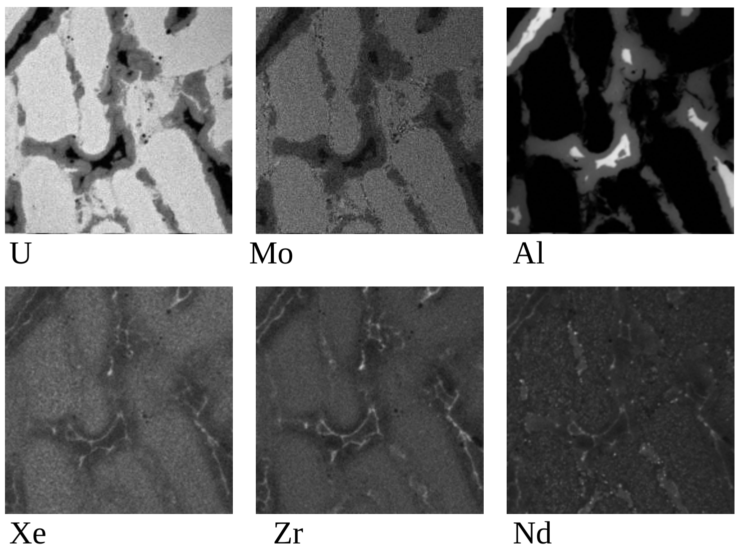


Fig. 7 : X-ray mappings of UMo7 plate meat (BU = 61%)

Then concentration profiles for the major phases existing in the irradiated fuel meat were determined by quantitative EPMA.

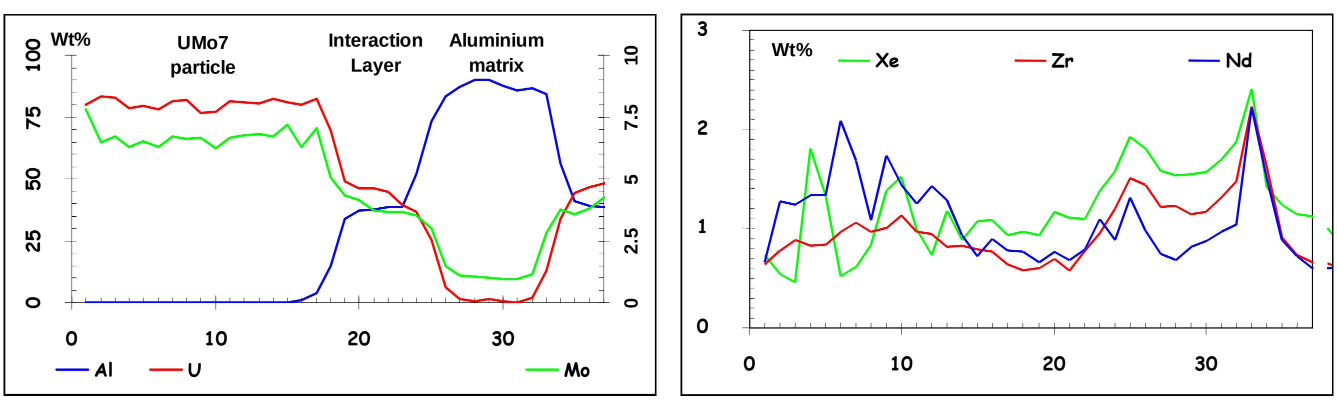


Fig. 8 : Concentration profiles from UMo7 particles to aluminium matrix, including interaction layer in between (BU=61%)

Fig. 8 gives the results for BU=61% location, From UMo7 particles centre to aluminium matrix.

These profiles show that :

- No aluminium diffusion is detected in the UMo particle
- The aluminium content in the interaction layer is not homogeneous. Nevertheless the global average compositions are found to be between "(U-Mo)Al_6" (BU= 61%) and "(U-Mo)Al_7" (BU= 67.5%). Moreover, a continuous decrease of the aluminium content in the interaction layer is observed from the aluminium side to the UMo side.
- Fission products accumulation at the interaction layer and aluminium matrix interface. However no porosity are detected at these interfaces. These accumulations could be induced by the reaction layer growth.
- Fission products implantation in the aluminium matrix.
- Local depletion of Xe is observed in the UMo particles due to the opening of bubbles during sample preparation. Therefore the neodymium quantitative measurements were used to estimate the local xenon creation, and the results indicate that only about 30% of the fission gas did not precipitate in the UMo particle bubbles. This result correlated to the bubble sizes analyses (see **Fig. 6**) leads to estimate the pressure in the bubbles at peak burnup location (67.5%) to roughly 7 MPa at room temperature.

5 Discussion

5.1 UMo Modelling

CEA has developed in close collaboration with ANL a thermo-mechanical code, called MAIA, for the modelling of the UMo fuel. MAIA uses a finite element method for the resolution of the thermal and mechanical problems. The physical models (interaction layer growth, FP swelling) are the ones used in the ANL code, PLATE [5]. Several correlations are available for the modelling of the growth of the oxide layer on the cladding.

The thermal modelling takes into account the volume fraction of each constituent of the meat that evolves in time. The thermal properties of the meat are then homogenized.

In MAIA, the swelling is the result of several phenomena : the reaction of the UMo with the aluminum matrix, fission product swelling of UMo, fission product swelling of reaction product and densification of the meat. The densification is assumed to balance the swelling phenomena : the global swelling of the meat only starts when the initial porosity has totally disappeared.

5.2 Meat composition

The interaction between UMo alloy particles and aluminium matrix is generally supposed to have a formulation of (U-Mo)Al_x with x =2, 3 or 4.4 by assuming a solid solution of Mo in the intermetallic compounds UAl_2, UAl_3 and $UAl_{4,4}$ [6].

Interaction compound in IRIS1 experiment is found to have a higher aluminium content, i.e. "(U-Mo)Al_6"" to "(U-Mo)Al_8". Such higher aluminium content interaction product has been recently observed [7] on heat treated fresh fuel at high temperature (550°C). However, in this last study the (U-Mo)Al_3 was still the predominant compound while after IRIS1 irradiation, at rather low temperature, it is not observed reaction compound with lower content than "(U-Mo)Al_6". Further investigations have to be done in order to understand the interaction compounds formation under irradiation. In particular the thermodynamic equilibrium of these phases with temperature and the effect of irradiation on their stability have to be investigated.

Moreover, it was supposed in previous studies [6] that a small amount of aluminium could be in dissolution into UMo fuel particles. So far, in IRIS 1, EPMA results do not show any aluminium in the

UMo7 alloy. For this point too, further investigations at higher temperature are needed to confirm the fact that there is no aluminium diffusion in UMo particles.
The modelling has been updated to include these observations : the compound of the interaction layer is taken as "(U-Mo)Al_7" and the diffusion model of aluminium into UMo particles is not activated. Under these conditions, MAIA gives the following volume fraction evolution of each meat constituent for the UMo7 plate at the metallographic sample n°2 location (see **Fig. 9**).

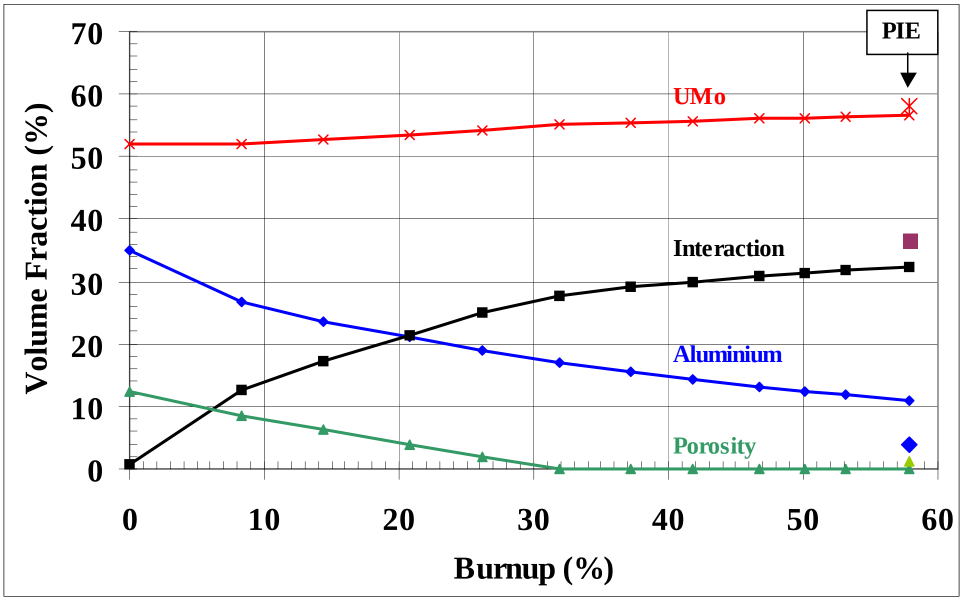


Fig. 9 : Volume fraction as calculated by MAIA code for sample n°2 (compare to PIE results)

The Table 4 gives a comparison of the MAIA code results with the measurements at the end of irradiation (PIE).

	Interaction layer thickness (µm)	UMo (%)	Interaction layer (%)	Al (%)	Porosity (%)
PIE	5.5	58	37	3.9	1.1
MAIA results	3.5	57	32	11	0

Table 4 : Modelling versus experiment at the end of irradiation on sample n° 2

The calculations are in rather good agreement with experimental data. However, improvements in the modelling are still necessary to get better predictions, especially for the aluminium depletion (remaining aluminium and interaction layer growth).

5.3 Swelling behaviour

The meat swelling versus the burnup is evaluated for each metallographic samples (see Fig. 10). It is deduced from IRIS1 thickness measurements with the following assumptions :

- Linear growth law of the plate surface oxide
- Oxide layer is supposed to be boehmite so when it growths it consumes half of its thickness of initial cladding
- The initial cladding thickness is equal to 380µm
- End irradiation gross gamma scans are representative of the burnup profile.

It can be seen that UMo7 and UMo9 exhibit roughly the same swelling behaviour. Moreover, within the investigated range of irradiation conditions, no dependencies with heat flux and temperature is observed on the meat swelling.

The graph given Fig. 10 shows two different behaviours as a function of the Burnup.

- For BU<30% (# 2 10^{27} fissions.m^{-3}), no apparent dimensional changes of the meat thickness.
- For BU>30%, the swelling rate growths linearly.

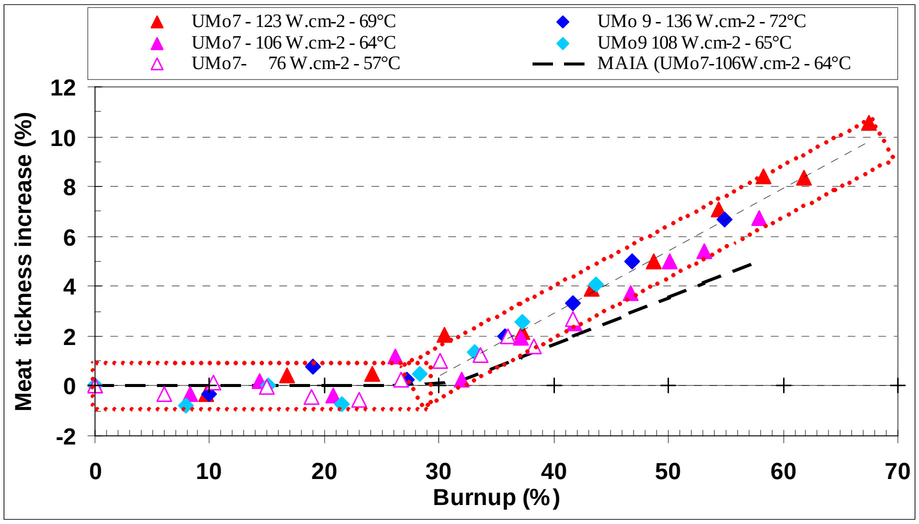


Fig. 10 : Meat thickness increase versus burnup for IRIS1 UMo7 and UMo9 plates at several locations

The modelling gives the total swelling of the meat and the different contributions of physical phenomena to it. (see Fig. 11). It appears that the total swelling is mainly driven by product fission accumulation in UMo particles and by densification.

For BU<30%, the swelling induced by interaction layer growth and fission product accumulation is accommodated by as-fabricated porosity. Therefore the total swelling is nil, as observed experimentally.

For BU>30%, the porosity has totally disappeared. The total swelling is then mainly driven by the fission product accumulation, bubble precipitation and growth in UMo particles.

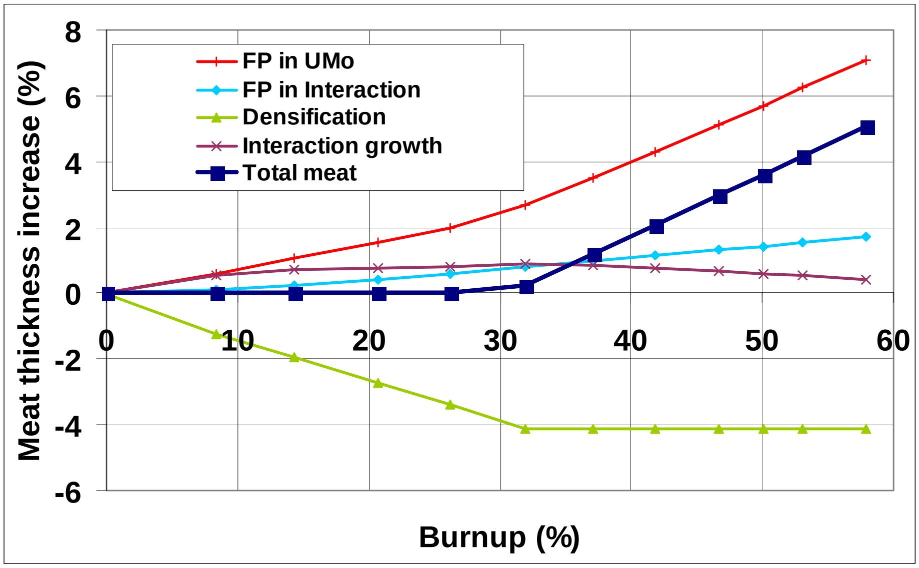


Fig. 11 : Fuel meat swelling components as calculated by MAIA code for sample n°2

The as-fabricated porosity seems to play a key role in the swelling behaviour of the plate by delaying it. It appears in these calculations that for this irradiation, the end of densification and the change in the swelling rate of UMo particles both occur around 30% burnup (2 10^{27} fissions.m^{-3}). It fits with the transition observed experimentally in plate thickness increase.
Some modelling improvements are needed to avoid the bias observed on the interaction growth evolution, here obtained as the difference between total swelling and the others contributions.

6 Conclusion

IRIS 1 irradiation and the wide set of PIE establish, up to 241 full power days of irradiation and a peak burn-up of 67.5%, the very good behaviour of high uranium loading UMo7 and UMo9 plates for a heat flux lower than 140 W.cm^{-2} and a BOL clad temperature under 75°C, due to :

- Very moderate plate swelling (less than 6%),
- And no tendency to abnormal voids interlinking.

The PIE done show that within the studied irradiation conditions:

- Fine distribution of little voids (0.5 µm mean diameter) inside the UMo particles at 67.5% burnup,
- Interaction product have a global formulation of "(U-Mo)Al_{-7}",
- There is fission products accumulation at the interaction layer and aluminium matrix interface,
- And there is no aluminium diffusion in the UMo fuel particles.

Finally, the modelling appears to be a useful tool to follow the UMo fuel behaviour under irradiation.

7 References

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